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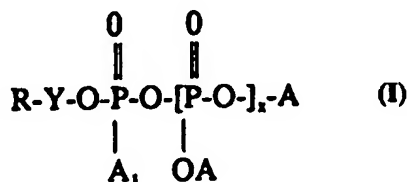
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(54) Title: SILICONE MODIFIED PHOSPHOLIPID COMPOSITIONS



(57) Abstract

A silicone-containing phospholipid composition having general formula (I), wherein: A is H, M or R-Y-; A<sub>1</sub> is H, OH, OM or R-Y-O-; M is a cation; X is a number from 0 to 5; Y is alkylene and R is a quaternized organosilicone amidoamine moiety wherein the organosilicone backbone is attached through at least one pyrrolidone containing amidoamine functional group.

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SILICONE MODIFIED PHOSPHOLIPID COMPOSITIONSRelated Applications

This application is a continuation in part of  
copen ding application Serial No. 420,748 filed April 12,  
5 1995.

Field of the Invention

The present invention relates to novel  
organosilicone compositions and, more particularly, to  
silicone compositions having a carboxyl functional group  
10 thereon and to derivatives thereof having at least one  
esterified phosphate group in the molecule.

Background of the Invention

Phosphate esters, quaternary ammonium compounds,  
betaines and certain substituted betaines are known in the  
15 art and have been commercially used over the years for a  
variety of applications, including those requiring  
surfactant properties. More recently, various betaine  
derivatives having, in general, specific quaternary  
compounds linked to phosphate esters referred to as  
20 phosphobetaines, and more particularly "synthetic  
phospholipids," have been disclosed, for example, in U.S.  
Patents 4,215,064, 4,233,192 and 4,380,637 to Lindemann et  
al.; U.S. Patents 4,209,449, 4,336,385 and 4,503,002 to  
Mayhew et al.; U.S. Patents 4,243,602, 4,283,542 and  
25 4,336,386 to O'Lenick et al; and U.S. Patent 4,617,404 to  
Lukenbach et al. These synthetic phospholipids are  
disclosed as exhibiting outstanding foaming, viscosity  
building, wetting, cleansing, detergency, anti-static,  
conditioning and emulsifying properties, making them useful  
30 in industrial applications calling for high performance  
surface active agents. The synthetic phospholipids are also  
described as being highly stable compounds which are well  
tolerated by human tissue (i.e. they exhibit exceptionally  
low oral toxicity and ocular irritation) and, hence, are  
35 well suited for use in a variety of personal care  
applications including cosmetic formulations as well as in  
industrial processes.

A variety of organosiloxane compositions including compositions which exhibit excellent properties as surface active agents, lubricants and the like are known and have been used commercially over the years, including for personal care and home care applications. In general, however, organosiloxane compositions are water insoluble, which has limited their use for many applications. Recently, particular types of betaine and phosphobetaine modified organosiloxanes having improved, although limited water solubility properties have been disclosed, for example, in U.S. Patents 4,609,750 and 4,654,161 to Kollmeier et al. and U.S. Patent 5,091,493 to O'Lenick et al. Such compositions are suggested as exhibiting high foaming characteristics in water, substantivity to a variety of surfaces and reduced irritation to the eyes and skin. While, as indicated, certain organosilicone containing phosphobetaine compositions and methods for preparing the same heretofore have been suggested, there has been no disclosure or suggestion of the novel carboxyl functional silicone compositions and silicone-containing phospholipid compositions of the present invention which compositions exhibit a wide range of properties including solubility in a variety of solvents, preferably water, and are useful for a variety of different applications.

#### Summary of the Invention

It is accordingly an object of the present invention to provide novel silicone-containing compositions which exhibit improved solubility characteristics particularly improved aqueous solubility.

It is another object of the present invention to provide novel silicone-containing phospholipid compositions which exhibit excellent surface-active properties including high foaming, are well tolerated by human tissue, are substantive to the surface of a variety of substrates such as fiber, and the like.

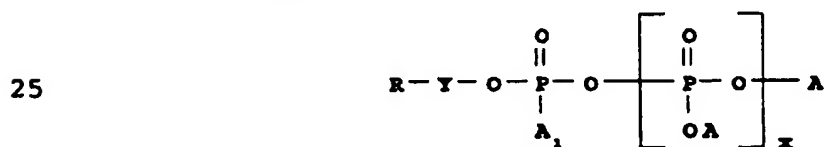
It is a further object of the present invention to provide novel soluble, preferably water soluble

silicone-containing phospholipid compositions having functional phosphorous containing groups linked terminally, laterally, or combination(s) of terminal and lateral linkages to the polysiloxane, which compositions can be prepared with a variety of concentrations of silicone, as desired or required.

It is still another object of the present invention to provide improved cosmetic and personal care preparations which include novel soluble, preferably water soluble, silicone-containing phospholipid compositions compatible with a variety of surfactant compositions and particularly compositions and methods for treating hair and for skin treatment.

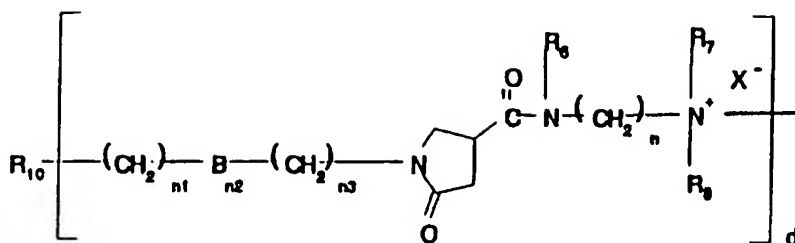
It is yet another object of the present invention to provide improved preparations useful in the manufacture and treatment of paper and textile products which include novel soluble, preferably water soluble, silicone-containing phospholipid compositions compatible with a variety of surfactant compositions.

In accordance with the present invention, there has now been discovered novel silicone containing phospholipid compositions that may be represented by the following general formula:



wherein:

- A is selected from H, M and R-Y-;
- A<sub>1</sub> is selected from H, OH, OM and R-Y-O-;
- x is 0 or an integer from 1 to 5;
- M is a cation, preferably an alkali metal;
- Y is a alkylene or substituted alkylene; and
- R is a quarternized organosilicone amidoamine moiety of the formula:



wherein:

$R_{10}$  is the silicone backbone chain to which a pyrrolidone-containing carboxyl or ester functional group or amidoamine derivative thereof as herein described can be attached as shown;

$R_6$  is hydrogen or alkyl, hydroxyalkyl or alkenyl of up to 6 carbon atoms, cycloalkyl of up to 6 carbon atoms, or polyoxyalkylene of up to 10 carbon atoms, preferably from 2 to 5 carbon atoms, within the oxyalkylene unit;

$R_7$  and  $R_8$ , which may be the same or different, are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl moiety and polyoxyalkylene of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the nitrogen to which they are attached may represent an N-heterocycle;

$R_9$ , which can be the same or different, is linear or branched alkylene of 1-12 carbon atoms;

$X^-$  is an anion, preferably a halogen;

$n$  is 0 or 2;

$n^1$  is 0 or 1;

$n^2$  is 0 or 1;

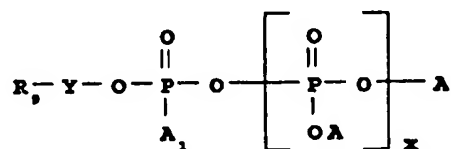
$n^3$  is an integer from 2 to 12;

$B$  is  $-NR_{11}$ , sulfur (S) or oxygen (O), wherein  $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ) with the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$

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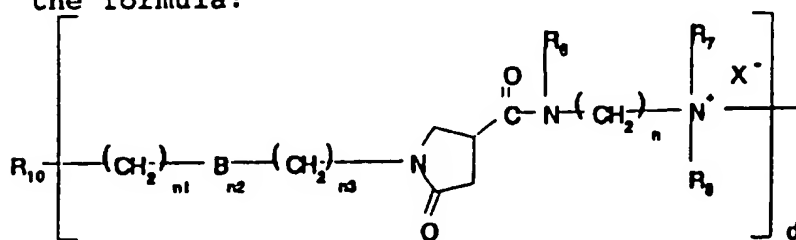
is 1, when n is 2 and n<sup>2</sup> is 1, n<sup>3</sup> is 0 or 1,  
and when n is 2 and n<sup>2</sup> is 0, n<sup>1</sup> is 0; and  
d is one or greater, preferably 2-10

In a further aspect of the present inventions  
5 there is provided novel silicone-containing phospholipid  
compositions that may be represented by the following  
general formula:



wherein:

- A is selected from H, M and R-Y-;  
10 A<sub>1</sub> is selected from H, OH, OM and R-Y-O-;  
M is a cation, preferably an alkali metal;  
x is 0 or an integer from 1 to 5;  
Y is alkylene or substituted alkylene; and  
R<sub>1</sub> is a quaternized silicone-containing  
15 amidoamine moiety of the formula a) and  
the mixture thereof with an organic  
amidoamine moiety of the formula b) or  
organic tertiary amine moiety of the  
formula c) or mixtures of the same:  
20 a) a quaternized organosilicone amidoamine moiety of  
the formula:



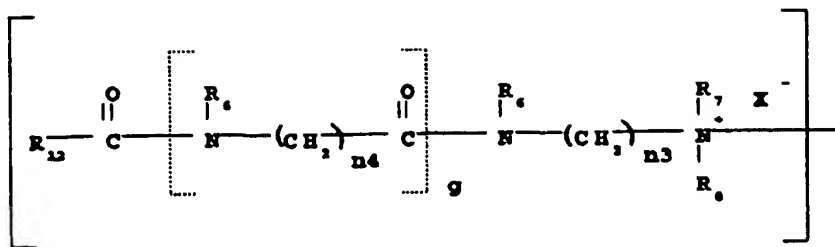
wherein:

R<sub>10</sub> is a silicone backbone chain to which  
a pyrrolidone containing carboxyl or

ester functional group or amido amine derivative thereof as hereinafter described can be attached;

- 5  $R_6$  is hydrogen or alkyl, hydroxyalkyl or alkenyl of up to 6 carbon atoms, cycloalkyl of up to 6 carbon atoms, or polyoxyalkylene of up to 10 carbon atoms, preferably from 2 to 5 carbon atoms, within the oxyalkylene unit;
- 10  $R_7$  and  $R_8$ , which may be the same or different, are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl, and polyoxyalkylene of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the nitrogen to which they are attached may represent an N-heterocycle.
- 15  $F$ , which can be the same or different, is linear or branched alkylene of 1-12 carbon atoms;
- 20  $X^-$  is an anion, preferably a halogen;  
 $n$  is 0 or 2;  
 $n^1$  is 0 or 1;  
 $n^2$  is 0 or 1;  
 $n^3$  is an integer from 2 to 12;
- 25  $B$  is  $-NR_{11}$ , sulfur (S) or oxygen (O) wherein  $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ); with the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$  is 1, when  $n$  is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1 and when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0; and
- 30  $d$  is one or greater, preferably 2-10;  
b) a quaternized organic amidoamine moiety of the formula:





wherein:

R<sub>12</sub> is alkyl, alkenyl, alkoxy or hydroxyalkyl of from 5 to 21 carbon atoms each, alkaryl or aryl of up to 20 carbon atoms;

5 R<sub>6</sub> is hydrogen or alkyl, hydroxyalkyl or alkenyl of up to 6 carbon atoms, cycloalkyl of up to 6 carbon atoms, or polyoxyalkylene of up to 10 carbon atoms, preferably of from 2 to 5 carbon atoms, within the  
10 oxyalkylene unit;

R<sub>7</sub> and R<sub>8</sub> which may be the same or different, are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl moiety, and polyoxyalkylene  
15 of up to 10 carbon atoms; in addition R<sub>7</sub> and R<sub>8</sub> taken together with the nitrogen to which they are attached may represent an N-heterocycle;

X<sup>-</sup> is an anion, preferably a halogen;

20 g is 0 or 1;

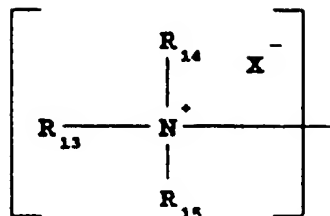
n<sup>3</sup> is integer from 2 to 12; and

n<sup>4</sup> is 1 or greater; and

c) an organic quaternized tertiary amine moiety of the formula:

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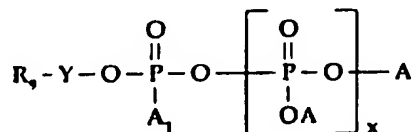


wherein:

$R_{13}$ ,  $R_{14}$  and  $R_{15}$  are the same or different and are alkyl, substituted alkyl, alkyl aryl or alkenyl groups of up to 16 carbon atoms with the proviso that the total carbon atoms in  $R_{13} + R_{14} + R_{15}$  is between 10 and 24; with the proviso that at least 5 equivalent weight percent and preferably to about 70 or 80 equivalent weight percent of the total equivalent weight of amine moieties of the phospholipid composition is a quaternized organosilicone amidoamine moiety.

It is evident from the general phospholipid formulae above that the functional phosphorus containing group(s) can be linked terminally, laterally or both terminally and laterally to the polysiloxane chain through the amidoamine and/or tertiary amine functional group.

In still a further aspect of the present invention there is provided a method of preparing a novel phospholipid composition that may be represented by the general formula:

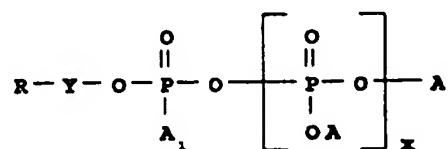


wherein

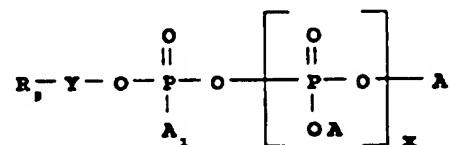
$A$ ,  $A_1$ ,  $Y$ ,  $M$ ,  $x$  and  $R_9$  are as described hereinabove which comprises reacting the combination of an organic silicone-containing amidoamine reactant with a polyphosphate, phosphite or phosphate ester halide reactant in the equivalent weight

ratios of from about 0.7 to 3.3 of total amidoamine and/or tertiary amine reactants to 1 of polyphosphate, phosphite or phosphate ester halide reactant until the amine reactant is completely reacted, with the proviso that at least 5 equivalent weight percent to about 70 or 80 equivalent weight percent of the total equivalent weight of amine reactants will be silicone containing.

In yet another aspect of the present invention there is provided cosmetic and personal care compositions comprising from about 0.1% to about 10%, preferably from about 1%, soluble, and preferably water soluble, silicone-containing phospholipid compositions selected from the group consisting of phospholipid compositions that may be represented by the general formula:



or that may be represented by the general formula:

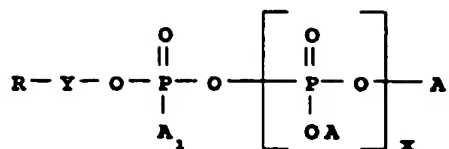


Wherein A, A<sub>1</sub>, R and R<sub>1</sub> are as defined above.

## 20 Description of the Preferred Embodiments

In accordance with the present invention, there are provided novel phospholipid compositions which comprise a class of silicone-containing phospholipid compositions represented by the general formulas:

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wherein:

A is selected from H, M and R-Y-;

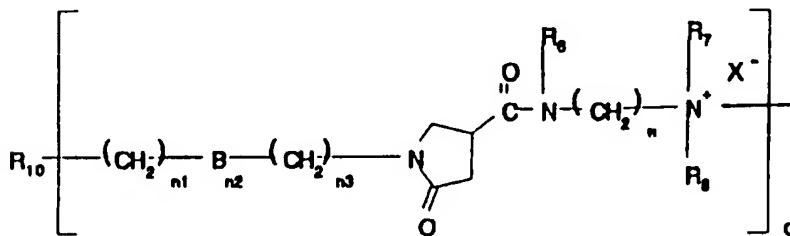
5 A<sub>1</sub> is selected from H, OH, OM and R-Y-O-;

x is 0 or an integer from 1 to 5;

M is a cation, preferably an alkali metal;

Y may be alkylene, optionally interrupted by  
up to 3 oxygen atoms, of up to 12 carbon  
atoms, which alkylene chain may optionally  
be substituted with lower alkyl,  
alkoxyalkyl or hydroxyalkyl, e.g. not more  
than 10 carbon atoms each; and

R is a novel quaternized organosilicone  
amidoamine moiety of the formula:



wherein:

R<sub>10</sub> is the silicone backbone chain to which at  
least one pyrrolidone containing carboxyl  
or ester functional group or amidoamine  
derivative thereof is attached as herein  
described;

R<sub>6</sub> is hydrogen or alkyl, hydroxyalkyl or  
alkenyl of up to 6 carbon atoms each,  
cycloalkyl of up to 6 carbon atoms, or  
polyoxyalkylene of up to 10 carbon  
atoms, preferably from 2 to 5 carbon  
atoms, within the oxyalkylene unit;

R<sub>7</sub> and R<sub>8</sub>, which may be the same or different,

are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl and polyoxyalkylene of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the N to which they are attached may represent an N-heterocycle.

F, which can be the same or different, is a linear or branched alkylene of 1-12 carbon atoms;

$X^-$  is an anion, preferably a halogen;

$n$  is 0 or 2;

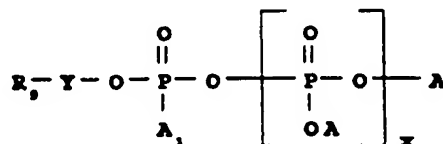
$n^1$  is 0 or 1;

$n^2$  is 0 or 1;

$n^3$  is an integer from 2 to 12;

B is,  $-NR_{11}$ , sulfur (S) or oxygen (O), wherein  $R_{11}$  is H or a lower alkyl ( $C_{1-6}$ ); with the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$  is 1, when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0 or 1 and when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0; and  $d$  is one or greater, preferably 2-10.

In an alternate embodiment of the present invention, there are also provided novel phospholipid compositions including high molecular weight polysiloxanes which comprise a class of silicone-containing phospholipid compositions which exhibit a surprising and unexpected solubility in aqueous systems and/or aqueous/co-solvent systems. Such novel silicone-containing phospholipid compositions may be represented by the general formula:



wherein:

A is selected from H, M and R-Y-;

$A_1$  is selected from H, OH, OM and R-Y-O-;

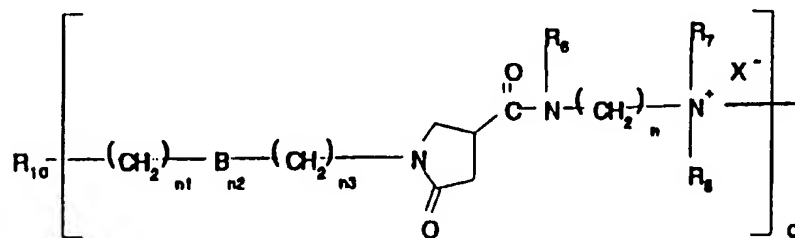
M is a cation, preferably an alkali metal;

x is 0 or an integer from 1 to 5;

Y may be alkylene optionally interrupted by up to 3 oxygen atoms, of up to 12 carbon atoms which alkylene chain may optionally be substituted with lower alkyl, alkoxyalkyl or hydroxyl, e.g. not more than 10 carbon atoms each; and

R<sub>9</sub> is a quaternized silicone-containing amidoamine moiety of the formula a) and the mixture thereof with an organic amidoamine moiety of the formula b) or tertiary amine moiety of the formula or mixtures of the same;

a) a quaternized organosilicone amidoamine moiety of the formula:



wherein:

R<sub>10</sub> is a silicone backbone chain to which at least one pyrrolidone containing carboxyl or ester functional group or amidoamine derivative thereof as herein described can be attached;

R<sub>6</sub> is hydrogen or alkyl, hydroxyalkyl or alkenyl of up to 6 carbon atoms each, or cycloalkyl of up to 6 carbon atoms, or polyoxyalkylene of up to 10 carbon atoms, preferably from 2 to 5 carbon atoms, within the oxyalkylene unit;

R<sub>7</sub> and R<sub>8</sub>, which may be the same or different,

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are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl, and polyoxyalkylene of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the N to which they are attached may represent an N-heterocycle;

$X^-$  is an anion, preferably a halogen;

$n$  is 0 or 2;

$n^1$  is 0 or 1;

10  $n^2$  is 0 or 1;

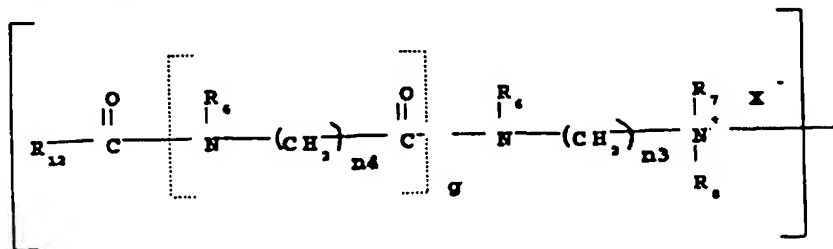
$n^3$  is an integer from 2 to 12;

B is  $-NR_{11}$ , sulfur (S) or oxygen (O), wherein  $R_{11}$  is hydrogen or lower alkyl; with the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$  is

15 1, when  $n$  is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1 and when  $n$  is 2 and  $n^1$  is 0,  $n^1$  is 0; and

$d$  is one or greater, preferably 2-10;

b) a quaternized organic amidoamine moiety of the formula:



20 wherein:

$R_{12}$  is alkyl, alkenyl, alkoxy or hydroxyalkyl of from 5 to 21 carbon atoms each, alkaryl or aryl of up to 20 carbon atoms;

25  $R_6$  is hydrogen or alkyl, hydroxyalkyl or alkenyl of up to 6 carbon atoms each, cycloalkyl of up to 6 carbon atoms, or polyoxyalkylene of up to 10 carbon atoms,

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preferably of from 2 to 5 carbon atoms,  
within the oxyalkylene unit;

$R_7$  and  $R_8$  which may be the same or different,  
are selected from alkyl, hydroxyalkyl,  
5 carboxyalkyl of up to 6 carbon atoms in  
each alkyl moiety, and polyoxyalkylene  
of up to 10 carbon atoms; in addition  $R_7$  and  
 $R_8$  taken together with the nitrogen to which  
they are attached may represent an N-  
10 heterocycle;

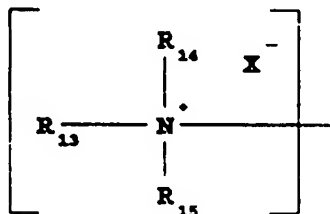
$X^-$  is an anion, preferably a halogen;

$g$  is 0 or 1;

$n^3$  is integer from 2 to 12; and

$n^4$  is 1 or greater; and/or

15 c) an organic quaternized tertiary amine  
moiety of the formula:



20 wherein:

$R_{13}$ ,  $R_{14}$  and  $R_{15}$  are the same or different and  
are alkyl, substituted alkyl, alkyl aryl or  
alkenyl groups of up to 16 carbon atoms  
with the proviso that the total carbon

25 atoms in  $R_{13} + R_{14} + R_{15}$  is between 10 and 24;

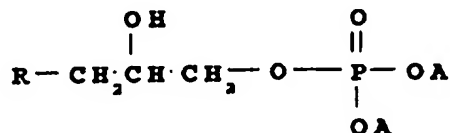
$X^-$  is an anion, preferably a halogen;

with the proviso that at least 5 equivalent  
weight percent and preferably to about 70 or  
80 equivalent weight percent of the total  
30 equivalent weight of amine moieties of the  
phospholipid composition is a quaternized  
organosilicone amidoamine moiety.

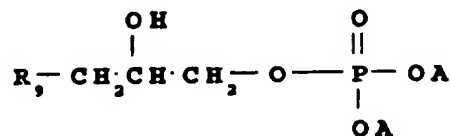


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Preferred silicone-containing phospholipid compositions of the invention wherein Y is 2-hydroxypropylene comprise a class of compositions which may be represented by the general formula;



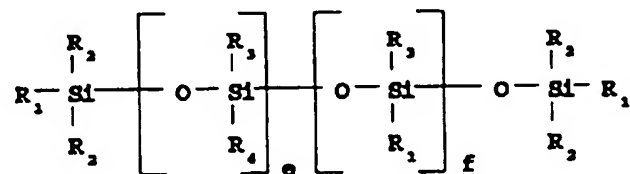
5 or



wherein

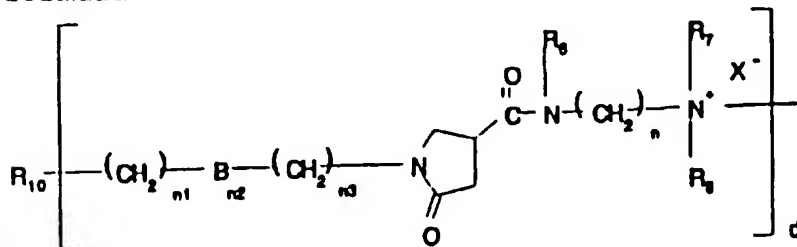
A, R and R<sub>2</sub> are as defined above.

The silicone backbone chain R<sub>10</sub> to which the pyrrolidone containing amidoamine functional group  
10 hereinabove shown (represented below as R<sub>1</sub>) are attached corresponds to the general formula:



wherein:

15 R<sub>1</sub>, which can be the same or different, can be selected from R<sub>2</sub>, a primary amine and a pyrrolidone containing group of the formula:



wherein at least one of  $R_1$  is a pyrrolidone containing amidoamine functional group as shown; F, which can be the same or different is a linear or branched alkylene of 1-12 carbon atoms;  $R_2$  is as defined below;  $R_7$  and  $R_8$  which may be the same or different are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the nitrogen to which they are attached may represent N-heterocycle; n is 0 or 2;  $n^1$  is 0 or 1;  $n^2$  is 0 or 1;  $n^3$  is an integer from 2 to 12; and B is  $-NR_{11}$ , sulfur (S) or oxygen (O), wherein  $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ); with the proviso that when n is 0 and  $n^2$  is 1,  $n^1$  is 1, when n is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1, and when n is 2 and  $n^2$  is 0,  $n^1$  is 0;

$R_2$  can be the same or different and can be selected from alkyl, aryl and olefinic (vinyl);

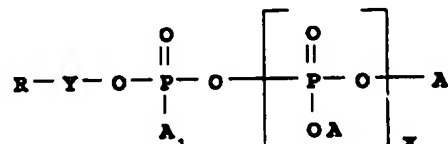
$R_3$  and  $R_4$ , which may be the same or different, are selected from alkyl, aryl, capped or uncapped polyoxyalkylene, alkaryl, aralkylene and alkenyl (vinyl);

e can be an integer from 0 to 50,000; and f can be an integer from 0 to 100.

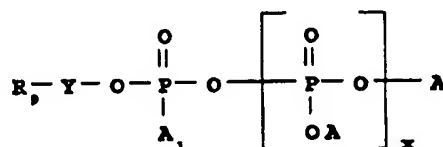
It is evident from the general formulae of the novel phospholipid compositions of the invention that the functional phosphorus containing group(s) can be linked terminally, laterally or both terminally and laterally to the siloxane chain through the amidoamine functional containing alkylene and/or alkylene containing heteroatom group.

In accordance with the invention, the phospholipid compositions of the invention can be prepared by reacting corresponding silicone containing amidoamine reactants or alternatively, combinations of silicone

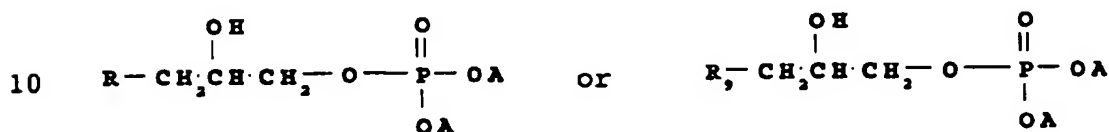
containing amidoamine and organic amidoamine and/or tertiary amine reactants with phosphate, polyphosphate or phosphite ester halide reactants in appropriate stoichiometric quantities as will be described in detail hereinafter to obtain the desired products of the formulae:



and



and preferably



wherein:

- A is as defined hereinabove;
- 15  $A_1$  is as defined hereinabove;
- Y is as defined hereinabove;
- x is as defined hereinabove;
- M is as defined hereinabove; and
- R is a quaternized organosilicone amidoamine moiety as defined hereinabove or,
- 20 alternatively,
- $R_p$  is a mixture of moieties selected from the

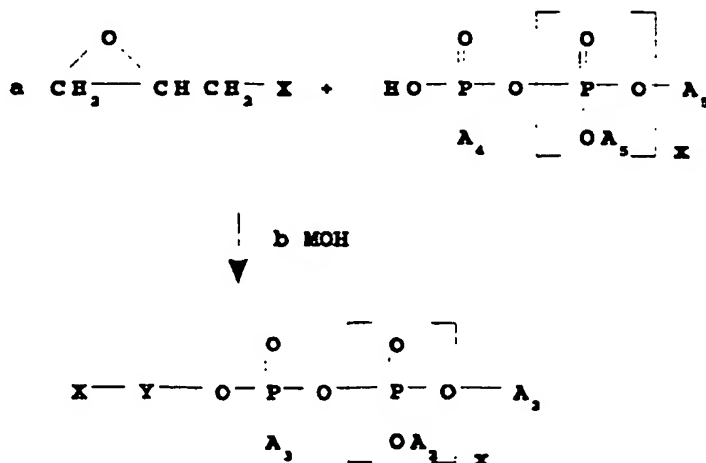
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group consisting of a quaternized  
 organosilicone amidoamine moiety,  
 quaternized organic amidoamine and/or  
 organic quaternized tertiary amine moiety  
 5 as defined hereinabove;  
 with the proviso that at least 5 equivalent  
 weight percent to about 70 equivalent  
 weight percent of the total equivalent  
 weight of amine moieties of the  
 10 phospholipid composition is a quaternized  
 organosilicone amidoamine moiety.

The intermediate reactants required in the  
 processes for preparing the silicone-modified phospholipid  
 15 compounds of the invention can be prepared as described  
 hereinafter.

Phosphate, polyphosphate and/or phosphite ester  
 intermediate reactants suitable for use can be prepared by  
 known procedures illustrated as follows:

20 I



25 wherein:

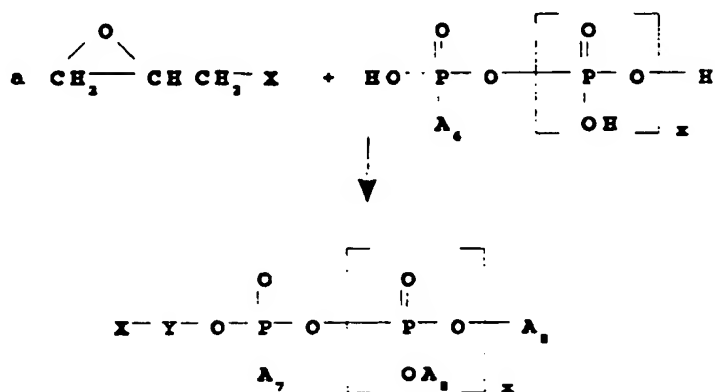
A<sub>2</sub> is selected from H, M and X-Y-;

19

- $A_3$  is selected from H, OH, OM and X-Y-O-;  
 $A_4$  is H, OM or OH;  
 $A_5$  is H or M;  
 a is from 0.5 to 3.5. preferably 1 to 3;  
 5 b is from 1 to 3; preferably 1 to 2;  
 M is a cation, preferably an alkali metal;  
 x is 0 or an integer from 1 to 5;  
 X is halogen; and  
 Y is 2-hydroxypropylene.

- 10 The above coupling reaction (I) is carried out in an aqueous media, preferably in the range of 30-50% concentration, having a pH range of 5.0 - 8.0.

II



- 15 wherein:  
 $A_6$  is selected from H or OH;  
 $A_7$  is selected from H, OH or X-Y-O-;  
 $A_8$  is H or -Y-X;  
 a is from 0.5 to 7, preferably from about 1 to 3;  
 20 x is 0 or an integer from 1 to 5;  
 X is halogen; and  
 Y is 2-hydroxypropylene.

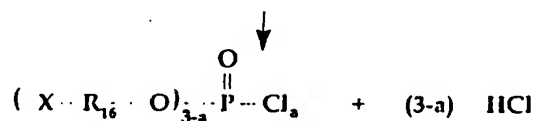
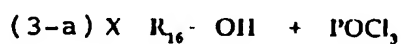
The above reaction (II) is preferably carried out in the absence of water.

- 25 Phosphate, phosphite and polyphosphate ester halide

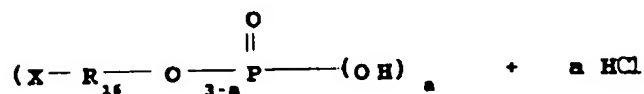
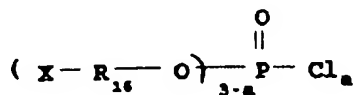
intermediate reactants for preparing phosphobetaine, pyrophosphobetaine and the like compositions of the invention can also be prepared by known procedures such as disclosed, for example, in U.S. Patent No. 4,617,414.

- 5 Also suitable as phosphate and phosphite intermediate halide reactants are such reactants prepared by known procedures illustrated as follows:

III (a)



10 III (b)



wherein:

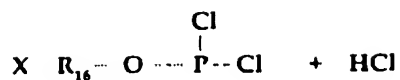
a is 0 or an integer from 1 to 2;

X is halogen; preferably bromine; and

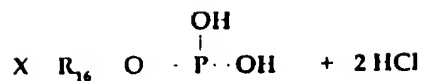
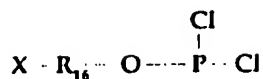
R<sub>16</sub> is alkylene.

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IV(a) IV (a)



IV (b)



wherein:

X is a halogen; preferably bromine; and

R<sub>16</sub> is alkylene.

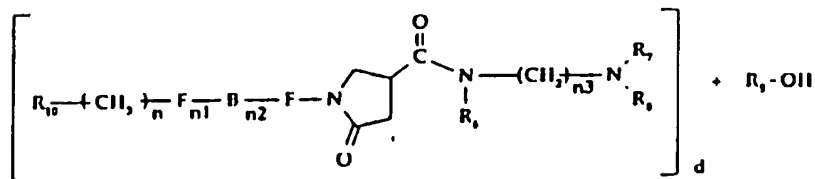
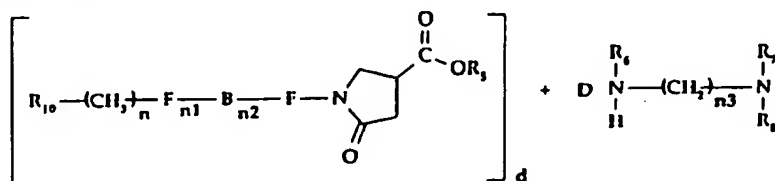
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Carrying out reactions III(a) and IV(b) in the presence of a tertiary amine HCl acceptor is preferred to prevent any secondary reaction with the generated HCl gas.

Silicone-containing amidoamines suitable for use as intermediate reactants in preparing the phospholipid compositions of the invention can be prepared as follows:

10

V



wherein:

- 5  $R_{10}$  is a silicone backbone chain to which at least one pyrrolidone containing carboxyl or ester functional group or amidoamine derivative thereof is attached as hereinabove shown;
- $R_5$  is hydrogen, lower alkyl ( $C_{1-6}$ ) or alkali metal;
- 10  $R_6$  is hydrogen or alkyl, hydroxyalkyl or alkenyl up to 6 carbon atoms each, cycloalkyl of up to 6 carbon atoms or polyoxyalkylene of up to 10 carbon atoms, preferably from 2 to 5 carbon atoms, within the oxyalkylene unit and at least one  $R_6$  is
- 15 hydrogen;
- $F$ , which can be the same or different is linear or branched alkylene of 1-12 carbon atoms;
- $R_7$  and  $R_8$ , which may be the same or different, are selected from alkyl, hydroxyalkyl,
- 20 carboxyalkyl of up to 6 carbon atoms in each alkyl and polyoxyalkylene of up to 10 carbon atoms; in addition  $R_7$  and  $R_8$  taken together with the nitrogen to which they are attached may represent an N-heterocycle.
- 25  $B$  is  $-NR_{11}$ , sulfur or oxygen wherein  $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ); with the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$  is 1, when  $n$  is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1 and when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0;
- 30  $n$  is 0 or 2;  
 $n^1$  is 0 or 1;  
 $n^2$  is 0 or 1;  
 $n^3$  is an integer from 2 to 12;
- $d$  and  $D$  is an integer from 1 or greater,
- 35 generally from 1-50 and preferably 2-10.  
The reactant ratio of the amine reactant to the carboxyl reactant on the silicon is



preferably 1:1 but can be varied in ratio  
from 1:0.8 to 1:1.2.

Silicone-containing amidoamines suitable for use  
as intermediate reactants are, thus, readily prepared by  
5 the above coupling reaction (V) from the novel polysiloxane  
compositions having one or more pyrrolidone containing  
functional carboxyl or ester group(s) as herein described.

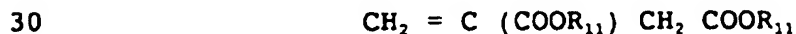
The above coupling reaction (V) for preparing the  
silicone-containing amidoamine intermediate can be carried  
10 out neat or can be carried out in an inert solvent such as  
xylene, toluene, chlorobenzene or the like. While the  
equivalent weight of suitable silicone-modified  
intermediate amidoamine reactants is not critical,  
preferably the equivalent weight of such reactants is from  
15 about 500 to 1500.

The novel carboxyl functional polysiloxanes or  
derivatives thereof (terminal, lateral or combinations of  
terminal and lateral) applicable for use in preparing the  
silicone-containing amidoamine intermediate reactants of  
20 the invention as set forth in the reaction sequence (V)  
illustrated above can be prepared by procedures such as  
disclosed in application Serial No. 420,746 filed April 12,  
1995. Such procedures include the reaction of corresponding  
silicone compositions or fluids having one or more  
25 functional primary amine groups with up to about one  
equivalent, preferably about stoichiometric quantities, of  
itaconic acid or its ester per functional amine group at an  
elevated temperature for the time sufficient for  
substantially all of the itaconic acid or its ester to  
30 react with the functional primary amine group(s).  
Preferably, from about 0.9 to about 1.1 equivalents of  
itaconic acid or its ester per functional primary amine  
group is reacted with the silicone fluid wherein  
substantially all itaconic acid is reacted with the  
35 functional primary amine group(s) and polysiloxane  
compositions with at least one pyrrolidone containing  
functional carboxyl group(s) and/or its ester are formed.

The reaction can be carried out neat or in an inert solvent such as alcohol, hydrocarbon solvent, chlorinated hydrocarbon and the like, in general, at elevated temperature, preferably from about 90°C to about 130°C. The reaction proceeds readily and generally complete reaction of the itaconic acid or its ester with the available functional primary amine groups and cyclization to form the pyrrolidone group will occur in from about 1 to 5 hours, with routine analytical techniques for amine and acid values as well as monitoring water and/or alcohol evolution being used to determine completion of the reaction.

Primary amine functional silicone fluids suitable for use having one or more primary amine functional group(s) linked terminally, laterally or both terminally and laterally, are well known and are available commercially, for example, from Dow Corning, Th. Goldschmidt AG and Shin-Etsu. While the equivalent weight of the silicone fluids or compositions which may be employed in the preparation of the pyrrolidone containing carboxyl functional polysiloxanes is not critical, and suitable compositions may have equivalent weights of 5,000 or even higher, silicone fluids having equivalent weights from about 500 to about 1500 are in general preferred.

As indicated, the polysiloxanes compositions are readily prepared by reaction of primary amine functional silicone fluids with itaconic acid or its ester. Itaconic acid (methylene succinic acid) is a compound of the formula:



wherein:

$\text{R}_{11}$ , which can be the same or different is hydrogen or lower alkyl ( $\text{C}_{1-6}$ ).

The compound is available commercially from Rhone Poulanc and Pfizer Chemicals Division and from Norflex, Inc., Greensboro, N.C. and is produced by known fermentation techniques although chemical synthesis methods  
5 are also known.

As noted above, silicone-containing phospholipid compositions of the invention can be prepared by reacting the desired silicone-containing amidoamine and phosphate, phosphite and/or polyphosphate ester halide reactants in  
10 appropriate desired stoichiometric proportions, in general, in molar equivalents of from about 0.7 to 3.3 of the amidoamine functional silicone reactant to 1 of the phosphate ester halide reactant. Such reaction can be carried out in a water solution or in conjunction with a  
15 co-solvent such as isopropyl alcohol, ethylene glycol, propylene glycol, ethyl cellosolve or the like. The reaction is carried out generally at elevated temperatures up to about 100°C, preferably from about 75° to 95°C, for a time ranging from about 1 to 5 hours, and generally until  
20 the amidoamine is substantially completely reacted. The course of the reaction can be determined by alkali number titration, ionic chloride determination, etc.

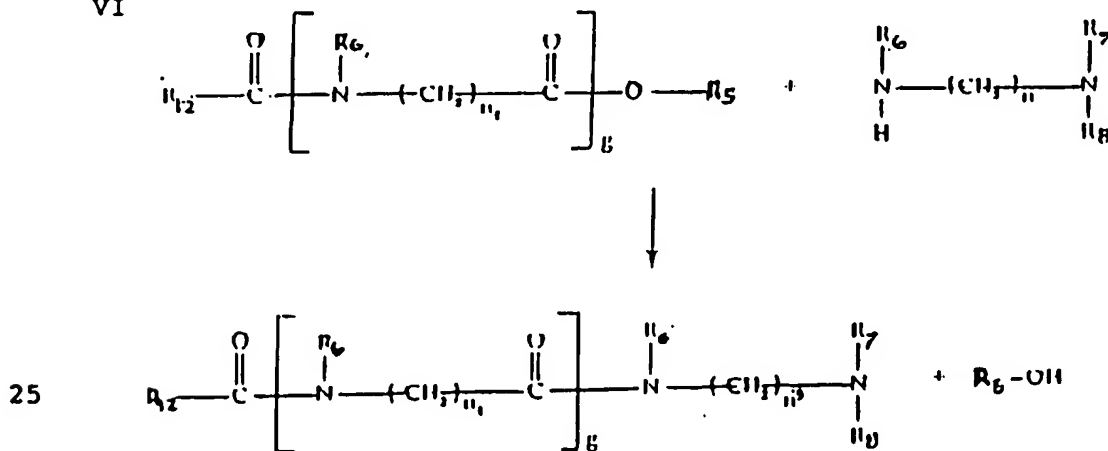
The reaction of the silicone-containing amidoamine and phosphate, phosphite and/or polyphosphate  
25 ester halide reactants can be readily carried out in an aqueous or an aqueous/organic co-solvent reaction system wherein the amine equivalent weight of silicone fluid reactant is in the range up to about 2,000. Reactions will go to completion as demonstrated by homogeneity of the  
30 reaction mixture. When the amine equivalent weight of the silicone reactant is greater than from about 1,200 to 2,000, the reactants are partially or completely insoluble in the reaction system and an incomplete reaction will result unless a co-solvent is used as referenced above.  
35 With silicone-containing amidoamine reactants having amine equivalent weights above about 2,000 to about 6,000, the addition of a co-solvent to an aqueous reaction system will

not increase the solubility of the reactants as evidenced by phase separation and/or other signs of incomplete reaction.

Surprisingly and unexpectedly, it has been found that alternate embodiments of the phospholipid compositions of the invention can be prepared in substantially completely soluble reaction systems using amidoamine functional silicone reactants as herein described having amine equivalent weights in excess of about 2,500 to about 6,000, or even greater, by also incorporating in the reaction system organic tertiary amine and/or preferably organic amidoamine reactants as herein described, in conjunction with the functional amidoamine silicone reactants. Suitable organic tertiary amine and/or amidoamine-containing reactants are added to the reaction system as a partial replacement of a substantially molar equivalent amount of the functional amidoamine silicone reactant, thus substantially maintaining the above noted molar equivalent ratios of amine reactants to phosphate ester halide reactants in the reaction mixture.

Organic amidoamine intermediate reactants suitable for use in preparing the phospholipid compositions of the invention can be prepared as follows:

VI



wherein:

$\text{R}_{12}$  is alkyl, alkenyl, alkoxy or hydroxyalkyl

of from 5 to 21 carbon atoms each alkaryl  
or aryl up to 20 carbon atoms;

5         $R_6$  is hydrogen or alkyl, hydroxyalkyl or  
alkenyl of up to 6 carbon atoms, cycoalkyl  
of up to 6 carbon atoms, or polyoxalkylene  
of up to 10 carbon atoms, preferably of  
from 2 to 5 carbon atoms, within the  
oxyalkylene unit;

10         $R_7$  and  $R_8$  is as hereinabove defined;

$R_9$  is hydrogen or alkyl;

$g$  is 0 or 1;

$n^3$  is an integer from 2 to 12; and

$n^4$  is 1 or greater.

15                The organic amidoamines suitable for use as  
intermediate reactants in preparing the phospholipid  
compositions are known and are generally prepared using  
conventional techniques such as shown in the above coupling  
reaction (VI). A variety of suitable amidoamines are  
20        commercially available as well as tertiary amino alkyl  
amines which are suitable for reaction with an acid or acid  
derivative to prepare suitable amidoamines. Suitable  
tertiary amino alkyl amines can be primary or secondary  
amines with the proviso that the total number of carbons in  
25        the acid portion of the molecule is greater than 6, i.e. to  
give a hydrophobic moiety necessary for surface activity  
properties. Suitable amidoamines may also be derived from  
acyl derivatives of aminoacid products such as glycine and  
sarcosine (N-methylglycine) including, for example,  
30        products available under the Tradename HAMPOSYL from the  
Hampshire Chemical Co.

Organic tertiary amine reactants suitable for use  
in preparing the phospholipid compositions of the invention  
can be prepared using procedures well known in the art and  
35        many suitable compositions are available.

Exemplary tertiary amines include:

tributylamine

bis(hydroxyethyl)hexylamine  
bis(2-hydroxyethyl)cocoamine  
N,N-dimethyl-dodecylamine  
N,N-dimethyl-tetradecylamine  
5 N,N-dimethyl-hexadecylamine  
N,N-dimethyl-cocoamine  
N,N-dimethyl-cetylamine  
dimethyl (C<sub>8</sub>-C<sub>16</sub>) alkyl amine  
N,N-dimethyl-octadecylamine

10 The reaction of a combination of organic tertiary amine and/or preferably organic amidoamine reactant(s) and silicone-containing amidoamine reactants with phosphate, phosphite and/or polyphosphate ester halide reactants will proceed to completion at an elevated temperature,  
15 preferably a temperature ranging from about 75°C to 95°C, with the formation of the silicone-containing phospholipid compositions of the invention. In a preferred embodiment, when using a silicone-containing amidoamine reactant having an amine equivalent weight in excess of about 4,000, a  
20 preferred organic amidoamine reactant may be an N-acylated amidoamine reactant derived from an amino acid or the mixture of a N-acylated amidoamine and an organic amidoamine reactant wherein the amount of N-acylated amidoamine used could be in excess of the stoichiometric  
25 amount of total amine reactants required.

The order of addition of the reactants is not critical although it may be advantageous to add the excess amount of N-acylated amidoamine reactant derived from an amino acid after all the other ingredients have been added.  
30 While a heterogeneous mixture may result when all reactants are admixed, the reaction system becomes homogeneous as the reaction proceeds. The reaction may start slowly while the mixture is heterogeneous but the reaction mixture will become substantially clear as the reaction proceeds. In  
35 accordance with the process of the invention, silicone-containing phospholipid compositions which contain at least 5 weight percent to about 70 or even 80 weight percent of

the silicone composition portion of the total solids of the reaction product can be prepared, which silicone-containing phospholipid compositions will be completely dispersible or soluble in an aqueous/solvent medium or, preferably, aqueous systems, while exhibiting surface active properties including low surface tension, high foaming and substantivity characteristics, low ocular and skin irritation and the like. Thus, it is possible by the combination of particular functional silicone-containing amidoamine reactants with organic amidoamine or tertiary amine reactants or mixtures of the same and phosphate ester halide reactants to obtain dispersible or soluble, preferably, aqueous dispersible or soluble silicone-containing phospholipid compositions which exhibit a wide range of beneficial properties suitable for use in a variety of applications.

The novel silicone-containing phospholipid compositions of the invention display many of the well known properties of silicones such as emolliency, emulsification, detackification, smoothing and surfactancy properties while, in addition, exhibiting the unique property of water-dispersibility or solubility and substantivity, thus making them easy to formulate into water-based systems and permitting them to provide long-lasting effects on various substrates such as skin, hair, natural and synthetic fibers (textiles), plastics, paper and the like. Moreover, the novel phospholipid compositions are non-irritating to the eyes and skin, are compatible with other surfactants as well as exhibiting a variety of other characteristics making them well suited for personal care and home care applications.

It is therefore a further aspect of the invention to use the novel compositions of the invention, for example, in cosmetic preparations and the like, especially in preparations for hair care and skin treatment. In this connection, personal care formulations can be shampoos, hair conditioners, skin treatment cremes and lotions and

after bath skin moisturizers, depending on whether the emphasis is on the cleansing effect or on the conditioning effect including the effect on better combability in hair care products. Other formulations where the unique properties of the novel compositions of the invention can be utilized include make-up cremes, sunscreens, lipstick, pressed powders, skin-toners, deodorants, antiperspirants and the like. Shampoos to which the novel phospholipid compositions of the invention have been added in amounts of 0.1 to 10 weight percent, and which contain up to about 30 weight percent of substances with detergent activity, besides water and possibly other additives cause washed hair to have fullness and an agreeable hand, to show a desirable gloss and to be easily combable. Practically no electrostatic charging of the hair is observed. In hair care products, such as, hair tonics or hair sprays, the composition of the invention in amounts of 0.1 to 5% by weight already bring about a significant improvement in combability of the hair as well as development of fullness and gloss.

Conventional additives, such as thickeners, perfumes, preservatives, complexing agents, foam stabilizers, opacifiers, luster development agents and the like may be added to the hair care products.

The novel phospholipid compositions may also be added to skin treating products. As a component of soap or skin cremes, they form a fine non-irritating, non-greasy film on the skin which does not evaporate and therefore provides long lasting protection. In addition, these novel phospholipid compositions also demonstrate useful emulsifying properties, including the unique capability of serving as emulsifying and even as solubilizing agents for a wide range of polysiloxanes.

Among other advantages of the novel phospholipid compositions of the invention are their usefulness as paper and textile sizing and lubricating agents; treating agents for fillers, paper and plastics; hydrophilic coatings for



glass, plastics and minerals; anticorrosion agents for metal products; adhesion promoters for metal primers and paints and emulsification agents and even as solubilizing agent for incorporation of a wide range of generally water insoluble silicone fluids in water based personal care and industrial compositions useful for many applications.

The preparation of specific compositions of the invention is illustrated by the following specific examples which are provided herein for purposes of illustration only and are not intended to limit the scope therein.

#### Example 1

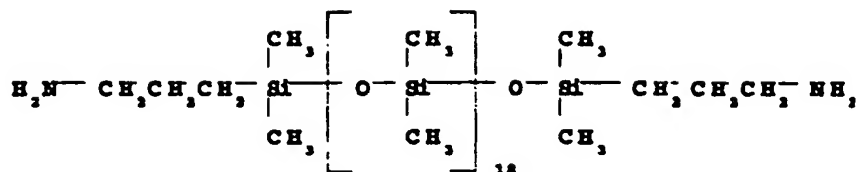
An alpha-omega bis primary amino alkyl dialkyl functional polysiloxane fluid obtained commercially under the designation Tegomer A-Si2120 from Goldschmidt Company is used in this example. The amine content of the fluid is 3.5% which corresponds to a molecular weight of 914.

91.4 grams of the above polysiloxane fluid (0.1 moles) is admixed with 26 grams (0.2 moles) of Itaconic Acid in a reaction vessel. Upon combination of the reactants, a heterogeneous mixture is formed. External heat is applied to the reaction vessel bringing the reaction mixture to a temperature of about 110°C, whereupon the reaction mixture becomes completely homogeneous while the temperature rises to 140°C.

After a heating period of 4 hours, a total of 7 1/2 ml. of volatiles are collected. The acid value of the reaction mixture is 81.6 (theoretical 95.5) while the alkali number is nil, thus confirming that there is the presence of carboxyl groups on the product.

#### Example 2

An alpha, omega-Bis primary amino alkyl dimethyl polysiloxane fluid with an average molecular weight about 1579.5 and having the general formula:



obtained commercially from Shin-Etsu under the designation X-22161A is used in this example.

A mixture of 994.5 grams of the above polysiloxane fluid (0.6296 moles) and 163.7 grams (1.25  
5 moles) of Itaconic Acid is formed in a reaction vessel and heated (slowly to about 90°C at which point an exotherm occurs raising the reaction vessel temperature to 130°C and water starts to evolve.

The reaction mixture is heated to and maintained  
10 at a temperature of 140°C to 150°C for a period of 3 hours during which time about 20ml. of water and other volatiles are collected. A clear, yellow viscous liquid is formed having an alkali number of 0.

The mixture is then cooled to 90°C and 192.6  
15 grams (an excess) of dimethylamino propylamine is admixed therewith. The temperature in the reaction vessel is increased to 170°C to 185°C and maintained at that temperature for an 4 additional hours during which time a total of 92 ml. of volatiles is collected.

The reaction mixture is then cooled to about 50°C  
20 and subjected to a vacuum of 30mm. While the vacuum is slowly drawn to 3mm, the reaction vessel temperature is increased from 75°C to 150°C. The product residue is then collected from the reaction vessel a 92% yield with an  
25 alkali number of 67 (theoretical alkali number is 57).

200.88 grams of the reaction product above (0.12  
moles) is then admixed with 75.4 grams of 40% active phosphate ester halide reactant and a 2:1 mixture of propylene glycol and water to obtain a solution having 30%  
30 solids. The phosphate ester halide reactant used is

prepared by the reaction of 3 moles of epichlorohydrin and one mole of sodium dihydrogen phosphate.

The reaction admixture is heated for 4 hours at a temperature range of 75°C to 85°C whereupon a homogeneous, clear liquid solution is obtained having a NaCl content of 1.8% (theoretical NaCl is 1.86%).

The product formed when mixed with water produced a great deal of stable foam whereas the polysiloxane functional amino fluid used as a starting material in the example provided no foam when mixed with water.

### Example 3

Another sample of a diamino polysiloxane fluid such as used in example 2 is used in this example. The average molecular weight of the sample is 1729 which corresponds to a percent amine of 1.85%.

To 43.2 grams (0.025 moles) of the above siloxane fluid in a reaction vessel is added with mixing 6.5 grams (0.05 moles) of Itaconic acid. The mixture is heated to 90°C whereupon an exotherm occurs raising the temperature to 130°C and resulting in a liquified viscous yellow mass. The reaction mixture is heated to and maintained at a temperature of 135°-140°C for 3 hours while some volatiles are collected and at which point the alkali number is zero.

There is then added to the reaction mixture 7.7 grams (0.075 moles, 33% excess) of dimethylamino propylamine (DMAPA) and the temperature is raised to 165°C where it is held for four additional hours. The reaction mixture is subjected to vacuum stripping to remove excess DMAPA at a reaction vessel temperature of 125°C and a vacuum of 10mm. The alkali number of the reaction product residue is 54 (theoretical 52.9).

A combination of 21.2 grams (0.01 moles) of the reaction product above, 6.25 grams (0.0061 mole) of 40% active phosphate ester halide reactant prepared as in example 2, 16 grams of isopropanol and 16 grams of water having a solid content of 40% is prepared in a reaction

vessel. The combined reactants are heated to a temperature of 95°C for four-five hours at which time a clear yellow solution is obtained having a NaCl content of 2.4% (theoretical 1.9%).

5           The product formed when added to water produces a great deal of stable foam.

#### Example 4

10           A pendant (lateral) amino functional silicone fluid having an average molecular weight of 4400 obtained from Shin-Etsu under the product designation KF865 is used in this example.

15           88 grams (0.02 moles) of the silicone fluid is admixed with 2.6 grams of Itaconic acid (0.02 moles) and heated to a temperature of 130°-140°C whereupon a clear melt is obtained and then continued heating for an additional two hours.

20           After heating for two hours, the reaction mixture is cooled to 70°C and 4.08 grams (an excess) of dimethylamino propyl amine (DMAPA) is admixed therewith. The reaction mixture is then heated to a temperature of 165°C for four hours, cooled to 70°C and vacuum stripped at 30-10mm for three hours while slowly raising the temperature to 110°C.

25           The reaction product residue is a clear liquid having an alkali number of 12.3 (theoretical 12.2) and an acid number of 0. I.R. analysis confirms the presence of an amide linkage.

#### Example 5

30           A pendant (lateral) aminofunctional silicone fluid obtained from Shin-Etsu under the product designation KF 865 is used in this example. The silicone fluid has an amine value of 0.2219 percent which corresponds to an amine equivalent weight of 5675.

35           665.9 grams (0.1173 equiv. wt.) of the silicone fluid and 15.25 grams of Itaconic acid (0.1173 mole) are combined with 150 ml of xylene in a reaction vessel and

heated to a temperature of 130° -140°C under reflux. After heating for 4 to 5 hours under reflux, 2.3 ml of water is removed (theory 2.1).

17.9 grams of dimethylpropyl amine (DMPA) is then admixed with the reaction mixture and heated under reflux at a temperature of 160° - 170°C for a period of about 4 hours during which time an additional 2.1 grams of water is removed. The reaction mixture is cooled to about 70°C and vacuum stripped at 30-40mm to remove low boiling volatiles. A vacuum of 5-10 mm is then applied to the reaction mixture and the reaction vessel is heated for three hours at 140°C. The reaction product is a clear liquid having an alkali number of 9 (theoretical 9.55) which corresponds to an amine equivalent weight of 6233.

264 grams (0.9428 moles) of cocoyl sarcosine obtained as HAMPOSYL C from Hampshire Chemical Corp. is reacted in another reaction vessel with 144 grams (an excess) of dimethylaminopropyl amine in 150ml of refluxing xylene under a nitrogen atmosphere, with water being removed as it is formed. After 4 hours, water no longer evolves and the xylene and any volatiles are removed by heating the reaction mixture to 130°C at 10mm vacuum for 3 hours. 348 grams of a product having an alkali number of 185 is obtained.

12.4 grams (0.002 equiv. wt.) of the above silicone/DMPA product are combined with 2.4 grams (0.008 moles) of the above sarcosine/DMPA product; 3.12 grams (0.0033 moles) of a 40% concentration of a phosphate ester halide reactant prepared by the reaction of 3 moles of epichlorohydrin with one mole of 85% phosphoric acid in the presence of one mole sodium hydroxide and 36 ml water in a reaction vessel. The reactant mixture is heated for 4 hours at a temperature of 85°-90°C. The reaction mixture is hazy and non-homogeneous. Upon the addition of 2.67 grams (0.009 mole) additional grams of the sarcosine/DMPA, the reaction mixture immediately becomes clear and homogeneous. The silicone content of the reaction product based on the total

solids is 64%. The product when added to water produces a great deal of stable foam.

#### Example 6

5 A mixture of the silicone/DMAPA and the 40% active solution of chlorohydroxy propyl phosphate reactants of example 5 when charged to a reaction vessel forms a completely non-homogeneous mixture even when heated at 100°C for several hours with only oily phases coating the reaction vessel.

10

#### Example 7

The silicone/DMAPA, cocoyl sarcosine DMAPA and phosphate ester reactants of example 5 are used in this example.

15 A combination of 18.7 grams (0.003 equiv.) of silicone/DMAPA; 2.12 grams (0.007 equiv.) of sarcosine/DMAPA; 3.12 grams of phosphate ester halide and 50 grams of water is formed in a reaction vessel and heated to 85-90°C for 3 hours. The reaction mixture becomes thick and difficult to agitate with only a small liquid phase.  
20 There is now added to the reaction mixture 20 grams of water and 5 grams (0.016 equiv.) of the cocoylsarcosine/DMAPA and the reaction mixture is heated for two additional hours. A homogenous, clear solution results having a silicone solid content of 69%.

25

#### Example 8

DiSodium 1, 3 Bis 3 chloro-2 hydroxy propyl pyrophosphate is prepared by charging 446 parts of  $\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$  (1 mole) and 178 parts  $\text{H}_3\text{P}_2\text{O}_7$  (1 mole) with 1000 parts deionized water to a reaction vessel and reacting the same  
30 with 320 parts epichlorohydrin at 60-80°C for 3-4 hours. 81.4 parts (0.2 equivalents) of the above reaction product is combined with a mixture of a pendant trimethylsilyl silicone pyrrolidone containing amidoamine having an

equivalent weight of 1845 (0.05 equivalent) and 57.6 parts of N-Dimethylaminopropyl linoleamide (0.15 equivalents) and then diluted with 480 grams of water to a 30% concentration. After the solution is adjusted to a pH of 8, the reaction mixture is heated to 90-95° for a period of 4-5 hours at which time a clear solution forms. The reaction is monitored via argentometric estimation for covalent chloride to ionic chloride and the reaction is completed in 5 hours.

The reaction product foams well in water.

#### Example 9

A 3-chloro-2 hydroxypropylester salt of phosphorous acid is prepared by charging 41 grams (0.5 mole) of phosphorus, 409 grams of water and 50 grams of 50% NaOH solution (0.6 mole) to a reaction vessel and warmed to 75°C. 46.25 grams (0.5 mole) of epichlorohydrin is then added and the reaction mixture is heated to 75°C for 1 1/2 hours with stirring.

196 parts of the combined epichlorohydrin-phosphite reaction mixture is admixed with 50% sodium hydroxide solution to achieve a pH of 8 followed by adding the combination of a pendant trimethylsilyl capped pyrrolidone containing silicone amidoamine having an equivalent weight of 3500 and 27.3 parts of cocoyl sarcosine amidoamine (0.09 equivalents). The reaction mixture is diluted with water to achieve 25% solids.

The reaction mixture is heated to 90°C for 2 hours whereupon a clear aqueous solution is formed.

#### Example 10

3-Bromopropyl diacid phosphate ( $\text{BrCH}_2\text{CH}_2\text{CH}_2\text{OPO}(\text{OH})_2$ ) is prepared by reacting 3-Bromopropanol with  $\text{POCl}_3$ , while utilizing one equivalent of triethylamine in a methylene chloride solvent. The

Dichloride is isolated and hydrolyzed to the diacid with water. The pH of the product in water is adjusted to 8 followed by the addition of an equivalent amount of a 50:50 combination of an alpha, omega, bistrimethylsilyl capped pyrrolidone carboxyl containing silicone amidoamine having  
5 an equivalent weight of 1845 and a N-(dimethylaminopropyl) derivative of linoleamide. The reaction mixture is adjusted with water to 30% of total solids.

The reaction mixture is heated for 4 hours at  
10 90°C. A clear solution is formed.

#### Example 11

The silicone/DMAPA and phosphate ester reactants of example 5 are used in this example.

A mixture of 24.92 grams (0.004 equivalents) of  
15 the silicone/DMAPA composition of example 5 (equivalent weight 6233) with 6.46 grams of N-(dimethylaminopropyl) linoleamide (molecular weight 404; 0.016 moles) is formed in a reaction vessel. To this mixture, 6.24 grams of the 40% active solution of chlorohydroxylpropyl phosphate of  
20 example 5 and 48 grams of water are added to prepare a 25% solution.

The reaction mixture is heated to 85-90°C for 4 hours. The reaction mixture goes through various stages from several immiscible phases, to a cloudy milky  
25 dispersion and then to a clear hazy phase. The sodium chloride content of the reaction product is 0.83% compared to a theoretical amount of 0.86%.

The reaction product containing 20 mole percent of silicone based on total amine requirements, when mixed  
30 with water produces a large amount of stable foam.

#### Example 12

A mixture of 37.39 grams (0.006 moles) of the silicone/DMAPA composition of example 5 and 5.6 grams of the N-(dimethylaminopropyl) linoleamide (mol. weight 404) of  
35 example 11 is mixed with 6.24 grams of the 40% active



solution of chlorohydroxy propyl phosphate ester reactant of example 5 and an additional 132 grams of water in a reaction vessel to prepare a 25% solution.

The reaction mixture is heated for 4 hours at 90°C and forms a hazy, single phase reaction product which is soluble in water and foams well.

The product has a sodium chloride content of 0.61% (0.64% theoretical) and a silicone content of 30 mole per cent.

10

#### Example 13

A reaction mixture of 12.4 grams (0.002 moles) of the silicone/DMAPA composition of example 5, 2.02 grams (0.005 moles) of the N-(dimethylaminopropyl) linoleamide of example 11, 0.7 grams (0.003 moles) of cocoyl dimethylamine and 3.12 grams of the 40% active solution of chlorohydroxylpropyl phosphate ester reactant of example 5 is charged to a reaction vessel. An additional 47 grams of water is added to the reaction mixture to form a 25% solution. The reaction mixture is heated for 4 hours at 90°C and a homogeneous solution is formed. The product is soluble in water and forms a stable foam.

20

#### Example 14

A reaction mixture of 7.4 grams (0.002 moles) of the silicone/DMAPA composition of example 5 (equivalent weight of 3715), 1.5 grams (0.005 moles) of soyadimethylamine (equivalent weight of 299) and 1.2 grams (0.003 moles) of the N-(dimethylaminopropyl) linoleamide (equivalent weight of 404) of example 11 (equivalent weight of 404) is prepared in a reaction vessel. 3.12 grams of the 40% active solution of chlorohydroxylpropyl phosphate ester reactant of example 5 and an additional 32 grams of water are added to the reaction mixture to form a 25% solution.

30

The reaction mixture is heated for 3 hours at 85° - 90°C and a clear solution is formed. The reaction product is soluble in water and forms of a great deal of stable foam.

35

Example 15

A Skin Care Creme composition is prepared having the following proportion of ingredients. The silicone-containing phospholipid composition of example 11 is used in this example.

<u>Part</u>	<u>Raw Material</u>	<u>Control A</u> <u>%Active</u>	<u>Test Sample</u> <u>%ACTIVE</u>	<u>Control B</u> <u>%Active</u>
A	Glycerin	10.0	10.0	10.0
A	Propylene Glycol			
10	Stearate	5.0	5.0	5.0
A	Cetyl Alcohol	2.0	2.0	2.0
A	PEG-20 Glycerol			
	Stearate	3.5	----	----
B	Silicone Phospho	----	----	----
15	lipid from Ex. 11			
B	Deionized Water	<u>76.5</u>	<u>79.5</u>	<u>80.0</u>
		100.0	100.0	100.0

Heat Parts A & B individually to 75°C. When hot, add Part B and Part B to Part A with agitation. Cool with mixing to 30°C.

Appearance after 24 hours at room temperature (25°C):

Control A - A moderate viscosity non-glossy, white creme

Control B - separated into layers

25 Test Sample - A moderate viscosity glossy white creme

Rubine Substantivity Test on wool exposed to creme and rinsed with warm water:

Control A - Non-substantive

Test Sample - Substantive

30 TRIALS OF CREME ON SKIN

When applied to skin, the test sample (contains phospholipid silicone) has a lighter textured, more easily spreadable consistency than the control, and when dry, after rub-in, it has a smooth, non-greasy emollient texture on the skin. The control A sample has a heavier texture and waxier feel on the skin when dry but was also emollient.

After rinsing the skin with warm water and drying, the test sample creme still exhibites residual emolliency and slip-smoothness while the control sample creme does not; this residual effect appears to be due to

the demonstrated substantivity of the silicone-containing phospholipid.

The silicone-containing phospholipid composition exhibits multi-functionality in that it serves as an emulsifier, an auxiliary emollient and it imparts emolliency that is water-resistant (a performance advantage for longer wearing skin care products and sunscreen formulations).

#### Example 16

The silicone-containing phospholipid compositions of examples 11, 13 and 14 are used in this example. Hair treating formulations such as shampoos and conditioners are prepared and evaluated as described below.

The evaluation is carried out as follows:

#### Procedures:

1. 1% by weight active silicone-containing solutions/dispersions are prepared in deionized water and adjusted with lactic acid to the 4-6 pH range.
2. Hair tresses weighing approximately 2 grams each are thoroughly shampooed, rinsed in water and then rinsed in isopropanol and dried prior to use.
3. The tresses are dipped in 50ml of the 1% active silicone-in-water dispersions, combed through ten times, drained and allowed to air-dry overnight prior to further evaluation.
4. A small R&D expert panel evaluated and ranked the tresses after treatment. A deionized water blank is included in the test as a performance benchmark. Numerical rankings assigned as follows:
  - 0 = Poor
  - 1 = Fair
  - 2 = Good
  - 3 = Very Good
  - 4 = Excellent
5. Silicone-containing compositions are also evaluated at 1.0% active level in 15% active Sodium Lauryl Sulfate

plus 2% active coconut monoethanolamide shampoo with pH adjusted to 4-6 range. After washing in the silicone-containing shampoos, hair samples are thoroughly rinsed in tap water, combed through and allowed to air dry prior to further evaluation.

Hair Tresses Washing in 1% Active Silicone in 15% SLS and 2% CMA Shampoo, Rinsed, Combed, Drained and Dried

1% Active Silicone in Shampoo	Shampoo Appearance	Wet Combing	Dry Combing	Total Cumulative Score
Example 11	Clear with only trace of opalescence	3.5	3.5	7.0
Example 13	Uniform, light opalescence	4.0	3.5	7.5
Example 14	Initially uniform light opalescence. On standing some supernatant layering	4.0	4.0	8.0
H <sub>2</sub> O	Clear	0	0	0

The example 14 sample exhibits excellent performance in combing characteristics and is slightly superior to the example 13 and example 11 samples, however, it is less soluble in this particular shampoo than the sample of example 11.

Hair Tresses Dipped in 1% Active Dispersions, Combed, Drained, and Dried with No Rinsing

5	1% Active Silicone Material Tested	Wet Combing	Hold of Dried Tress	Dry Combing	Texture vs. Control	Total Cumulative Score
10	Example 11 in water	3.0	1.0	3.0	3.0	10
	Example 13 in water	3.0	1.0	3.5	4.0	11.5
15	Example 14 in water	4.0	1.0	4.0	4.0	13
	H <sub>2</sub> O Blank	0	0	0	0	0

20 While the best performance of the water-based systems is exhibited by the preparation containing the phospholipid composition of example 14, formulations containing the phospholipid compositions of examples 11 and 13 exhibit advantageous hair treating characteristics.

Example 17

25 The tissue paper softening of the silicone-containing phospholipid composition of example 11 is evaluated in this example.

Procedure:

- 30 1. 1% by weight active silicone solution/dispersion is prepared in deionized water and adjusted with lactic acid to the 4-6 pH range.
- 35 2. Evaluation is conducted using 8" x 9" paper tissues weighing approximately 1.5 grams each. The lower half of each tissue is dipped briefly into the 1% active silicone solution being tested, then the tissues are withdrawn, allowed to drain, dried and equilibrated for several hours at ambient temperature and humidity. A deionized water blank is included in the test regimen.

3. Treated tissues are evaluated (undipped versus dipped portions) and ranked for softness by a small R&D expert panel. Numerical softness rankings were assigned as follows:

- 5           0 = Poor/harsh textures  
             1 = Fair  
             2 = Good  
             3 = Very Good  
             4 = Excellent/very soft textures

10   Paper Tissues Evaluated for Softness/Texture After Dipping in 1% Active Silicone Dispersion Followed by Draining and Drying

<u>1% Active Silicone Material Tested</u>	<u>Softness/Texture</u>
Example 11 sample in water	3.5
15   Deionized water blank	0

The tissue softening characteristics is exhibited by the formulation prepared with the silicone-containing phospholipid composition of example 11.

Example 18

20           The silicone-containing phospholipid composition of example 11 is used in this example. The compatability of the silicone-containing phospholipid composition with a shampoo containing an anionic detergent is evaluated.

25   Testing for Compatability of a Cationic Silicone Derivative with Anionic Shampoo

30           To test for the compatability of the cationic silicone containing phospholipid in typical anionic shampoo systems, an all-ammonium shampoo system often used in conditioning shampoos is prepared with a 15% by weight "hole" in the formulation to allow for addition of the silicone-containing phospholipid composition to material to determine if the system would remain clear. The formula for this shampoo formulation is shown below.

45

All-Ammonium Shampoo with 15% "Hole"

	<u>% "Active"</u>	<u>% "As Is"</u>
NH <sub>4</sub> Lauryl Ether Sulfate Colonial ALS-HV (28%)	13.0	46.4
5 NH <sub>4</sub> Lauryl Sulfate Sipon L-22 (28%)	7.0	25.0
Coconut Monoethanolamide MONAMID CMA	3.0	3.0
10 Ammonium Xylene Sulfonate Witconate NXS (40%)	2.0	5.0
Citric Acid	Q.S to pH 6.25	Q.S to pH 6.25
"Hole"	15.0	15.0
Demineralized Water	Q.S	<u>5.6</u>
		100.0%

15 It was found that the shampoo formulation remains clear even after the addition of 15% by weight "as is" (4.5% silicone active) silicone-containing phospholipid composition of example 11. The shampoo also shows a noticeable drop in viscosity and a marked conditioned feel

20 on the skin after rinsing and drying versus the same shampoo with no silicone addition.

In a further evaluation a commercially marketed shampoo (a clear system) is mixed with 1% and 5% by weight active levels of the cationic silicone-containing phospholipid of example 11 in water to check for its

25 compatability in the anionic shampoo system.

In combination with the commercial shampoo, formulations prepared with both 1% and 5% added silicone levels remained clear after addition to the system. The

30 shampoo with 5% active silicone added also shows a noticeable drop in viscosity, while the 1% active silicone shampoo shows little change. From these studies it appears that this class of cationic silicone containing phospholipid compositions is compatible with clear anionic

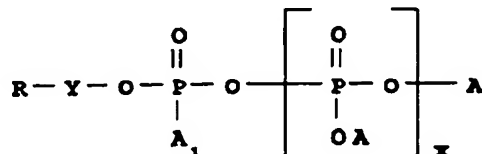
35 shampoo systems, even at rather high levels.

Although some preferred embodiments have been described, many modifications and variations may be made thereto in light of the above teachings. It is therefore to be understood that within the scope of the appended claims,  
5 the invention may be practiced otherwise than as specifically described and illustrated.



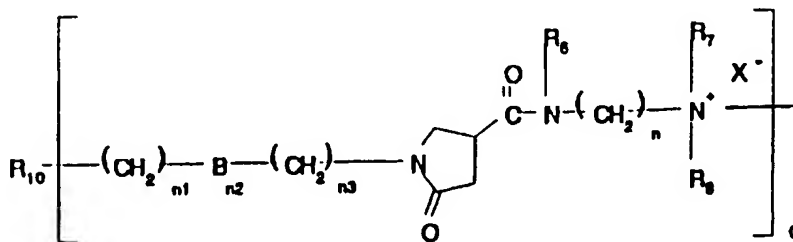
WHAT IS CLAIMED IS:

1. A silicone-containing phospholipid composition having the general formula:



wherein:

- 5 A is selected from H, M or R-Y-;  
 A<sub>1</sub> is selected from H, OH, OM or R-Y-O-;  
 M is a cation;  
 Y is substituted or unsubstituted alkylene  
 optionally interrupted by up to  
 10 3 oxygen atoms;  
 x is 0 or an integer from 1-5; and  
 R is a quaternized organosilicone amidoamine  
 reactant moiety of the formula:



15 wherein:

- R<sub>10</sub> is the silicone backbone chain to which at  
 least one pyrrolidone containing amidoamine  
 functional group is attached;  
 R<sub>6</sub> is hydrogen or alkyl, hydroxyalkyl or  
 20 alkenyl of up to 6 carbon atoms each,  
 cycloalkyl of up to 6 carbon atoms, or

polyoxyalkylene of up to 10 carbon atoms within the oxyalkylene unit;

$R_7$  and  $R_8$ , which may be the same or different, are selected from alkyl, hydroxyalkyl, carboxyalkyl of up to 6 carbon atoms in each alkyl moiety or polyoxyalkylene of up to 10 carbon atoms or in addition  $R_7$  and  $R_8$  taken together with the nitrogen to which they are attached represents an N-heterocycle;

F which can be the same or different is linear or branched alkylene of 1-12 carbon atoms;

$X^-$  is an anion;

$n$  is 0 or 2;

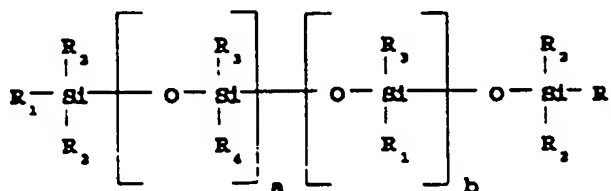
$n^1$  is 0 or 1;

$n^2$  is 0 or 1;

$n^3$  is an integer from 2 to 12.

B is  $-NR_{11}$  sulfur or oxygen, wherein  $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ); with the proviso that when  $n^1$  is 0 and  $n^2$  is 1,  $n^1$  is 1, when  $n$  is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1 and when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0; and;  $d$  is one or greater.

2. The silicone-containing phospholipid composition as claimed in claim 1, wherein  $R_{10}$  is a polysiloxane backbone chain to which at least one pyrrolidone containing amidoamine functional group is attached, said polysiloxane backbone chain having the formula:



wherein:

$R_1$  can be the same or different and is selected from  $R_2$ , a primary amine or a pyrrolidone containing carboxyl functional group or amidoamine derivative thereof; with the proviso that at least one of  $R_1$  is a pyrrolidone containing amidoamine functional group.

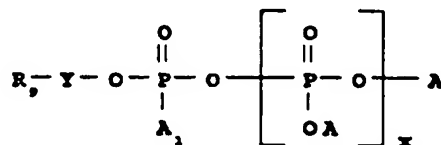
$R_2$  can be the same or different and is selected from alkyl, aryl or olefinic;  
 $R_3$  and  $R_4$ , which may be the same or different are selected from alkyl, aryl, capped or uncapped polyoxyalkylene, alkaryl, aralkylene or alkenyl;  
 $e$  is an integer from 0 to 50,000;  
 $f$  is an integer from 0 to 100;

3. The silicone-containing phospholipid composition as claimed in claim 2, wherein  $f$  is 0.

4. The silicone-containing phospholipid composition as claimed in claim 2, wherein the terminal groups  $R_1$  are  $R_2$  and  $e$  and  $f$  are each at least 1.

5. The silicone-containing phospholipid composition as claimed in claim 1, wherein  $M$  is an alkali metal,  $X^-$  is a halogen, and  $d$  is an integer from 2 to 10.

6. Silicone-containing phospholipid compositions having the following general formula:

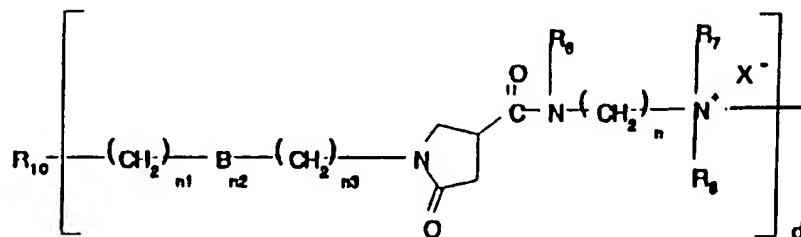


wherein:

$A$  is selected from H, M or  $R_2-Y-$ ;  
 $A_1$  is selected from H, OH, OM or  $R-Y-O-$ ;  
 $M$  is a cation;  
 $x$  is 0 or an integer from 1 to 5;

Y is alkylene or substituted alkylene; and  
 R<sub>9</sub> is a mixture of a quaternized silicone-  
 containing amidoamine moiety of the formula  
 a) and organic amidoamine of the formula b)  
 5 or organic tertiary amine moiety of the  
 formula c) or mixtures of the same.

a) a quaternized organosilicone amidoamine moiety of  
 the formula:



wherein:

10 R<sub>10</sub> is a silicone backbone chain to which at  
 least one pyrrolidone containing amidoamine  
 functional group is attached;

R<sub>6</sub> is hydrogen, alkyl, hydroxyalkyl, alkenyl of  
 up to 6 carbon atoms each, cycloalkyl of up  
 15 to 6 carbon atoms, or polyoxyalkylene of up  
 to 10 carbon atoms within the oxyalkylene  
 unit;

R<sub>7</sub> and R<sub>8</sub>, which may be the same or different,  
 are selected from alkyl, hydroxyalkyl,  
 20 carboxyalkyl of up to 6 carbon atoms in  
 each alkyl or polyoxyalkylene of up to 10  
 carbon atoms or in addition R<sub>7</sub> and R<sub>8</sub> taken  
 together with the nitrogen to which they  
 are attached represent an N-heterocycle;

25 X<sup>-</sup> is an anion;

n is 0 or 2;

n<sup>1</sup> 0 or 1;

n<sup>2</sup> is 0 or 1;

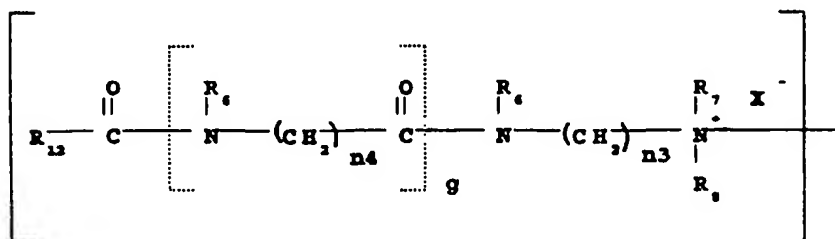
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$n^3$  is an integer from 2 to 12;

B is  $-NR_{11}$ , sulfur (S) or oxygen (O), wherein  
 $R_{11}$  is hydrogen or lower alkyl ( $C_{1-6}$ ); with  
 the proviso that when  $n$  is 0 and  $n^2$  is 1,  $n^1$   
 5 is 1, when  $n$  is 2 and  $n^2$  is 1,  $n^1$  is 0 or 1  
 and when  $n$  is 2 and  $n^2$  is 0,  $n^1$  is 0; and  
 $d$  is one or greater;

b) a quaternized organic amidoamine moiety  
 of the formula:

10



wherein:

$R_{12}$  is alkyl, alkenyl, alkoxy or hydroxyalkyl  
 of from 5 to 21 carbon atoms each, alkaryl  
 or aryl of up to 20 carbon atoms;

15  $R_6$  is hydrogen, alkyl, hydroxyalkyl or  
 alkenyl of up to 6 carbon atoms each,  
 cycoalkyl of up to 6 carbon atoms, or  
 polyoxyalkylene of up to 10 carbon atoms  
 within the oxyalkylene unit;

20  $R_7$  and  $R_8$  which may be the same or different,  
 are selected from alkyl, hydroxyalkyl,  
 carboxyalkyl of up to 6 carbon atoms in  
 each alkyl moiety or polyoxyalkylene  
 of up to 10 carbon atoms or in addition  $R_7$   
 25 and  $R_8$  taken together with the nitrogen to  
 which they are attached may represent an N-  
 heterocycle;

$X^-$  is an anion;

52

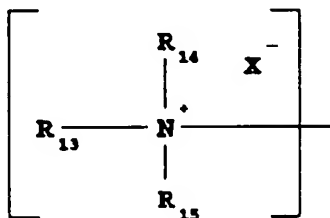
g is 0 or 1;

$n^3$  is integer from 2 to 12; and

$n^4$  is 1 or greater; and

c) an organic quaternized tertiary amine moiety of the formula:

10



wherein:

$R_{13}$ ,  $R_{14}$  and  $R_{15}$  are the same or different and are alkyl, substituted alkyl, alkylaryl or alkenyl groups of up to 16 carbon atoms with the proviso that the total carbon atoms in  $R_{13} + R_{14} + R_{15}$  is between 10 and 24;

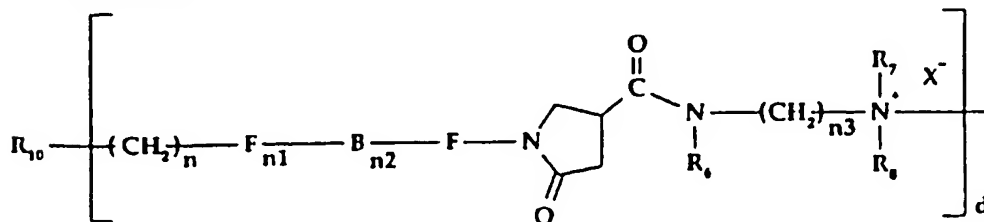
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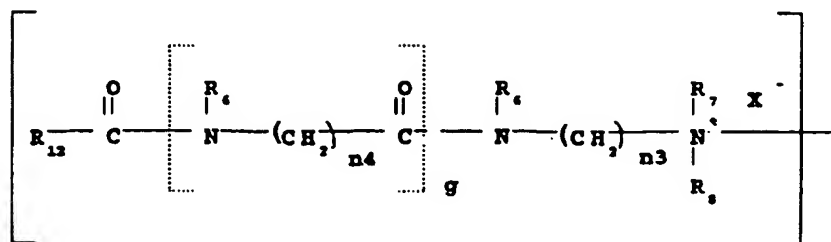
with the proviso that at least 5 equivalent weight percent to about 80 equivalent weight percent of the total equivalent weight of amine moieties of the phospholipid composition is a quaternized organosilicone amidoamine moiety.

7. The silicone-containing phospholipid compositions as claimed in claim 6, wherein  $R_9$  is a mixture of;

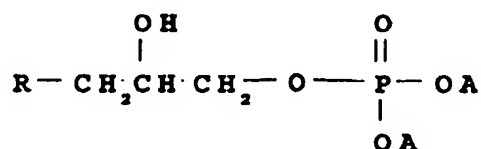
a) quaternized organosilicone amidoamine moieties of the formula:



b) quatermized organic amidoamine moieties of the formula:



8. The phospholipid compositions as claimed in claim 7, wherein  $\text{R}_{10}$  is a silicone backbone chain to which at least one pyrrolidone containing amidoamine functional group is attached, said polysiloxane backbone chain having the formula:



wherein:

10  $\text{R}_1$  can be the same or different and is selected from  $\text{R}_2$ , a primary amine or a pyrrolidone containing carboxyl functional group or amidoamine derivative thereof; with the proviso that at least one of  $\text{R}_1$  is a  
15 pyrrolidone containing amidoamine functional group;

$\text{R}_2$  can be the same or different and is selected from alkyl, aryl or olefinic;

20  $\text{R}_3$  and  $\text{R}_4$ , which may be the same or different are selected from alkyl, aryl, capped or uncapped polyoxyalkylene, alkaryl, aralkylene or alkenyl;

$e$  is an integer from 0 to 50,000;

$f$  is an integer from 0 to 100;

9. The phospholipid compositions as claimed in claim 6, wherein g in the quaternized organic amidoamine moiety b) is 1.

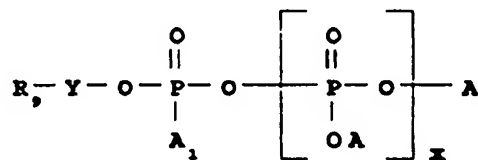
10. The phospholipid composition as claimed in claim 7, wherein g in the quaternized organic amidoamine moiety b is 1.

11. The phospholipid compositions as claimed in claim 8, wherein f is 0.

12. The phospholipid compositions as claimed in claim 8, wherein the terminal groups R<sub>1</sub> are R<sub>2</sub> and e and f are each at least 1.

13. The phospholipid compositions as claimed in claim 8, wherein R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> are each methyl.

14. A method of preparing phospholipid compositions represented by the general formula:



wherein:

A is selected from H, M or R<sub>g</sub>-Y-;

A<sub>1</sub> is selected from H, OH, OM or R<sub>g</sub>-Y-O-;

M is a cation;

20 x is 0 or an integer from 1 to 5;

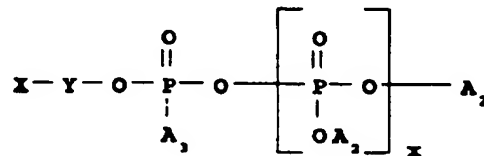
Y is alkylene or substituted alkylene; and

R<sub>g</sub> is a mixture of quaternized amidoamine and/or tertiary amine moieties as claimed in claim 6;

25 which comprises reacting the combination of an organic amidoamine and/or organic tertiary amine reactant and a silicone-containing pyrrolidone-containing amidoamine reactant with a phosphate, phosphate or polyphosphate ester reactant in the equivalent weight ratios of from about 0.7 to 3.3 of total amidoamine and/or tertiary amine reactants  
30 to 1 of phosphate, phosphite or polyphosphate ester reactant



until the amine reactant is substantially completely reacted, with the proviso that at least 5 equivalent weight percent to about 70 equivalent weight percent of the total equivalent weight of amine reactants will be silicone containing, said phosphate ester reactant being of the general formula:



wherein:

A<sub>2</sub> is selected from H, M or X-Y-;

A<sub>3</sub> is selected from H, OH, OM or Y-O-;

10 M is a cation;

Y is alkylene or substituted alkylene;

X is halogen; and

x is 0 or an integer from 1 to 5.

15 15. The silicone-containing phospholipid composition as claimed in Claim 1, wherein Y is 2 hydroxy propylene.

16. The silicone-containing phospholipid composition as claimed in Claim 1, wherein Y is substituted with lower alkyl, alkoxy, alkyl with not more than 10  
20 carbon atoms each or with hydroxy.

17. The silicone-containing phospholipid composition as claimed in Claim 6, wherein Y is 2 hydroxy propylene.

18. A cosmetic composition for the care of skin  
25 and hair comprising from about 0.1% to about 10% by weight of a silicone-containing phospholipid composition as claimed in Claim 2.

19. A cosmetic composition for the care of skin  
and hair comprising from about 0.1% to about 10% by weight  
30 of a silicone-containing phospholipid composition as claimed in Claim 6.

20. A cosmetic composition for the care of skin and hair comprising from about 0.1% to about 10% by weight of a silicone-containing phospholipid composition as claimed in Claim 3.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US96/04989

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(6) :C08G 77/06, 77/26; C07F 7/02; A61K 7/06

US CL :528/25, 26, 38; 548/406; 424/70.122, 78.03, 78.36

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 528/25, 26, 38; 548/406; 424/70.122, 78/03, 78.36

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US, A, 5,093,452 (O'LENICK, JR.) 03 March 1992, see entire document.	1-12
A	US, A, 2,843,615 (LINVILLE) 15 July 1958, see entire document.	1-12
A	US, A, 3,441,537 (LENGNICK) 29 April 1969, see entire document.	1-12
A	US, A, 4,093,641 (PLUEDDEMANN) 06 June 1978, see entire document.	1-12



Further documents are listed in the continuation of Box C.



See patent family annex.

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\*Z\*

document member of the same patent family

Date of the actual completion of the international search

22 MAY 1996

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